Occurrence and distribution of pesticide residues in selected seasonal vegetables from West Bengal

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ABSTRACT

A study was conducted on selected vegetables from West Bengal to assess the level of organochlorine pesticide residues and the concentration of ∑OCPs was ranged between, <0.01–65.07 µg kg^{-1} with average of 9.67±2.34 µg kg^{-1} (wet wt.). The concentration of ∑DDT, ∑HCH, aldrin, dieldrin and heptachlor was 3.49±0.93 µg kg^{-1}, 2.07±0.53 µg kg^{-1}, 1.32±0.65 µg kg^{-1}, 1.36±1.18 µg kg^{-1} and 1.80±0.4 µg kg^{-1} (wet wt) respectively. Isomer composition was determined from the observed concentrations to identify the possible contamination sources. Ratio of α-HCH/γ-HCH varied in the range of 0-5.69 with mean value of 0.76 which reflects the usage of technical HCH and lindane. The ratio of p,p'-DDT/∑DDTs, DDTs/∑DDT, DDE+DDD)/∑DDT, and DDT/DDE was 0.30, 0.50, 1.17 and 1.97 respectively, indicating DDT contaminations from biotransformation and transported depositions. The observed residue levels of OCPs were below maximum residue limits (MRLs) indicating minimal risk to the consumers.

Keywords: Pesticides, DDT, HCH, aldrin, dieldrin, heptachlor, vegetables.
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INTRODUCTION

Organochlorine pesticides (OCPs) are toxic compounds with tendency to persist in the environment, and their physico-chemical characteristics make them bio-accumulative in nature [1-2]. OCPs have a wide range of acute and chronic health effects, including cancer, neurological damage, reproductive disorders, immune suppression, birth defects and suspected endocrine disruption [3-4].

Organochlorine pesticides have been widely used in public health and agriculture production in developed and developing countries including India. India is the fourth largest pesticide producer in the world after US, Japan and China. The production of pesticides in India is approximately 85

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TMT (thousand metric tonnes), about 50 TMT of this quantity is used annually, and insecticides alone account for 71% of this consumption [5-6]. The consumption of pesticides in Indian agriculture is comparatively low (0.5 kg/ha), (only 3.75% of global consumption) as compared to 12.0, 7.0, 6.6, and 3.0 kg.ha^{-1} in Japan, USA, Korea and Germany, respectively [7]. Indiscriminate use of pesticides in agriculture leads to accumulation in consumable vegetables.

Vegetables are indispensable components of Indian diet both in terms of quantities consumed and nutritional value as majority of Indians are vegetarian, with per capita consumption of 135 g per day. Thus the information on residue levels of OCPs in vegetables is very important for the human health perspective, as the dietary intake being the main non-occupational route of exposure to organochlorines [8]. Contamination of vegetables with pesticides residue has been reported worldwide including India [9-19].

In West Bengal, 4100 MT of the technical grade pesticides were applied to 5.123 million hectares of agricultural land during 2004-05. Several studies on organochlorine pesticides in different matrices including food commodities have been reported from West Bengal [20-26].

This paper presents the results of a study carried out on chlorinated pesticides (HCHs, DDTs, aldrin, dieldrin and heptachlor) in selected vegetables from markets in Kolkata, West Bengal, India. The observed concentrations of organochlorine pesticides were further compared with the Recommended Maximum Residual Levels (MRLs) proposed by Indian government [27] and European commission [28].

MATERIALS AND METHODS

Sampling
Cauliflower, radish, tomato, carrot, spinach, and eggplant samples were collected from local vegetable markets of south Kolkata for analysis. The vegetables were collected in clean polyethylene bags, labelled; ice preserved, transported to the laboratory and was kept in refrigerator till further extraction.

Sample Extraction
Samples were washed with deionised distilled water, dried on filter paper, cut into small pieces with the help of grater and mixed thoroughly. Twenty grams of each vegetable was grinded with 10-15 g anhydrous sodium sulphate in warring blender. The grinded sample was extracted with 50 ml acetone on mechanical shaker for one hour. The acetone extract was filtered by employing vacuum suction and the process was repeated three times for complete extraction. The filtrate was concentrated to about 50 ml using Rotatory Vacuum evaporator (Buchi, Germany) and subjected to liquid-liquid partitioning with hexane in separatory funnel. Hexane layer containing pesticides residue was collected passing through anhydrous sodium sulphate. Aqueous phase was again subjected to hexane extraction (three times) for leftover residues. Pooled hexane fractions were concentrated to about 10 ml.

Sample Extract Clean-Up
Concentrated hexane extracts were passed through glass column containing activated charcoal and anhydrous sodium sulphate to clean the pigment contents. The florisil column chromatography was

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performed to remove other interfering aliphatic compounds. Briefly a glass column (300 mm x 30 mm) was packed from bottom up with 25 g activated florisil (60-100 mesh) and 5.0 g anhydrous sodium sulphate. The column was pre-rinsed with 50 ml n-hexane before loading the sample extract, elution of analytes was subsequently carried out using 170 ml hexane and the collected elute was concentrated to 2.0 ml. The concentrated extract was transferred to sample vial and 2 µl was injected onto a gas chromatograph equipped with an electron capture detector (GC-ECD) for quantification.

Instrumental Quantification

Identification and quantification of pesticide compounds was carried out using gas chromatograph (Varian Star 3400cx, Australia) equipped with ^{63}Ni electron capture detector (ECD). Separation of OCP compounds was accomplished using a fused silica capillary column (RTX-5) with 30 m x 0.25mm ID and 0.5 µm of stationary phase (5% diphenyl-95% dimethyl polysiloxane). The column oven temperature program was initially maintained at 170 ^{0}C, increased @ 7 ^{0}C min^{-1} to 220 ^{0}C and again ramped to 250 ^{0}C @ 5 ^{0}C min^{-1} and finally held for 7.0 min. The injector and detector temperature were maintained at 250 ^{0}C and 350 ^{0}C respectively. Purified Nitrogen was used as carrier gas at a flow rate of 1.0 ml min^{-1}.

Analytical Quality Control

Certified reference standard solutions from Sigma-Aldrich, USA were used for calibration of the instrument. The identification of the separated compounds was conducted by accurate matching of retention times with those of the corresponding standards. Resolved peaks were integrated using Varian Star workstation software and the concentrations of target compounds were determined by external standard method using the response factors from the five level calibration curves of the standards and multiplying to the peak areas of the samples. Appropriate quality control (QC) measures were performed, including analysis of procedural blanks to check the cross contamination and interferences (analyte concentrations were <MDL 'method detection limit'), random duplicate samples (Standard deviation <5), matrix spiked samples, calibration curves with the r^2 value of >0.999, calibration verification (standard deviation ±5-10%). Recoveries from spiked samples were in the range of 72-111 (±6-12) for studied pesticide compounds. The recoveries were satisfactory and the results were not corrected for the recovery. Each sample was analysed in duplicate and the average values were used in calculations. The results of the analysis are reported as wet weight (wet wt.) basis in µg kg^{-1}. A reporting limit of > 0.01 µg kg^{-1} wet wt. was set for further calculations. Levels falling below reporting limit or below MDL (<0.01 µg kg^{-1} wet wt.) were taken as zero (0) in the final compilation.

RESULTS AND DISCUSSION

Distribution of OCPs in Vegetables

The OCPs monitored were o,*p'*-DDE, o,*p'*-DDD, *p,p'*-DDD, *o,p'*-DDT and *p,p'*-DDT (DDTs), α-HCH and γ-HCH (HCHs), aldrin, dieldrin and heptachlor. The overall pesticide occurrence was in the order as: ∑DDT (3.49 µg kg^{-1}) > ∑HCH (2.07 µg kg^{-1}) > heptachlor (1.80 µg kg^{-1}) > dieldrin (1.36 µg kg^{-1}) > aldrin (1.32 µg kg^{-1}) (Table 1).

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Table 1: Concentration of chlorinated pesticides in vegetables (µg·kg⁻¹ wet wt)

Pesticide	Mean	Minimum	Maximum	Std error	MRLs Indian	EC
α-HCH	0.56	<0.01	1.55	0.09	-	-
γ-HCH	1.66	<0.01	13.53	0.50	-	-
∑HCH	2.07	<0.01	14.59	0.53	1000	50
DDT	1.76	<0.01	12.09	0.53	-	-
DDD	0.77	<0.01	11.32	0.37	-	-
DDE	1.04	<0.01	6.29	0.29	-	-
∑DDT	3.49	<0.01	25.67	0.93	3500	50
Aldrin	1.32	<0.01	18.31	0.65	100	10
Dieldrin	1.36	<0.01	34.11	1.18	100	10
Heptachlor	1.80	<0.01	7.81	0.34	50	10
∑OCPs	**9.67**	**<0.01**	**65.07**	**2.34**	**-**	**-**

DDT, DDE and DDD are sum of o,p' and p,p' isomers

Pesticide contamination in selected vegetables was in the order; radish > cauliflower > eggplant > tomato > carrot > spinach (Figure 1). The highest concentration of ∑HCH, ∑DDT, aldrin, dieldrin and heptachlor was 2.45 ±1.17 µg kg⁻¹, 8.52±3.96, 1.31±0.78, 5.79±1.58 and 5.88±2.44 µg kg⁻¹, respectively in radish. However, lowest concentration of ∑HCH, ∑DDT, aldrin, dieldrin and heptachlor was 0.60±0.36 (µg·kg⁻¹) in carrot, 1.24±0.61 (µg·kg⁻¹) in spinach, 0.24±0.12 (µg·kg⁻¹) in eggplant, 0.07±0.02 (µg·kg⁻¹) in spinach and 0.73±0.36 (µg·kg⁻¹) again in spinach.
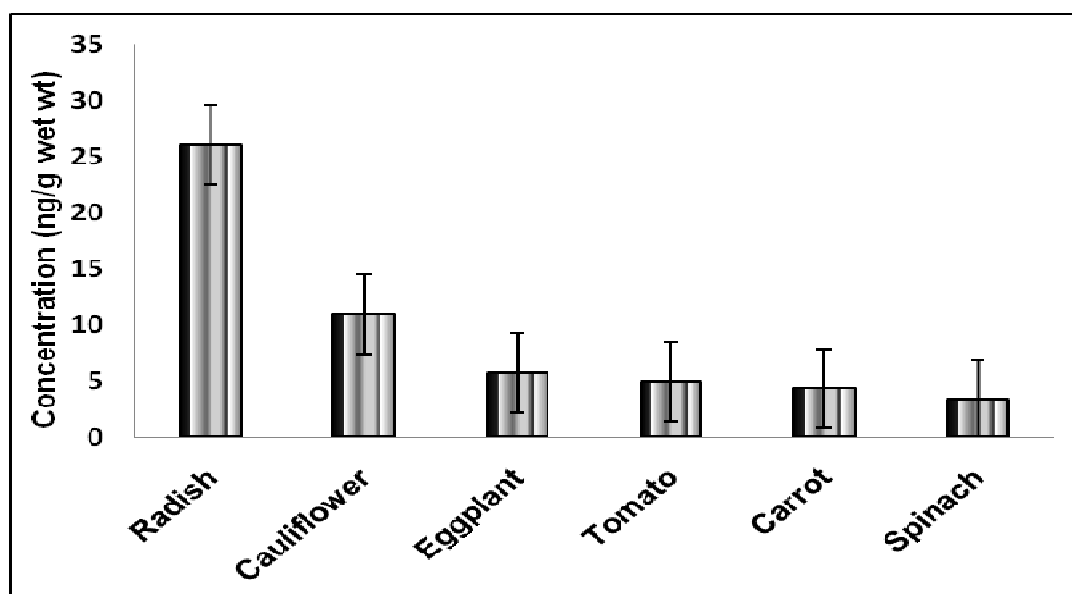

Figure 1: Total pesticide concentrations in selected vegetables

DDTs alone accounted for 23% of the total OCP contaminations in vegetables, followed by HCHs (14%), heptachlor (11%), aldrin and dieldrin (8% each) (Figure 2). The concentrations of pesticide compounds in different vegetables are presented in Table 2 & 3. Radish is the most and eggplant is least contaminated among all the studied vegetables with 24.00±6.60 µg·kg⁻¹ and 2.92±0.70 µg·kg⁻¹ respectively, of total OCPs. Vegetables in this study had residue levels of OCPs far much below the

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recommended maximum residue limits (MRLs) set by European Commission and Indian government, indicating minimal risk to the consumers.

Possible source of OCPs

Differences in composition of isomers in the environment could indicate different contamination sources [29]. Technical HCH has been used as broad spectrum pesticides for agricultural purposes, which has been banned since 1997 in India.
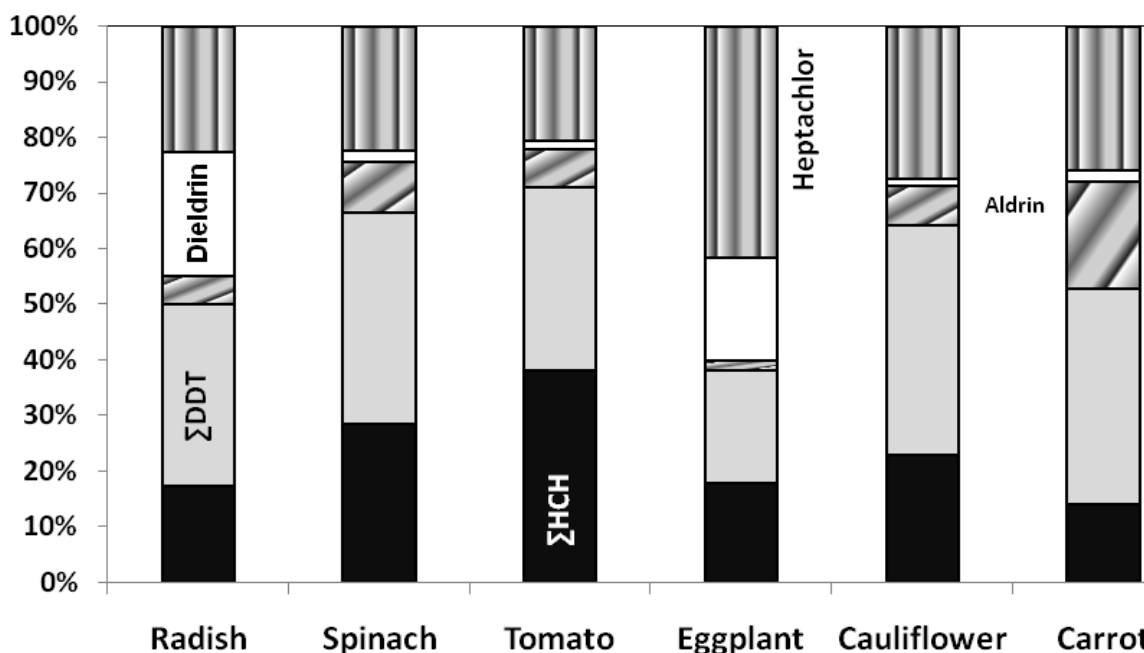

Figure 2: Accumulation of pesticides (in %) in vegetable samples

Technical HCH consists principally four isomers, α-HCH (60-70%), β-HCH (5-12%), γ-HCH (10-15%), δ-HCH (6-10%), while lindane contains >99% of γ-HCH [30]. The ratio of α-HCH to γ-HCH has been used to identify the possible HCH source. The ratio of α-HCH to γ-HCH between 4 and 7 is indicative of fresh input of technical HCH [31]. However, a lindane source show the reduced ratio close or <1 [32]. Ratio of α-HCH/γ-HCH varied in the range of 0-5.69 with mean value of 0.76 (Table 4), reflecting the usage of technical HCH as well as lindane in this study area. Studies anticipated the use of technical HCH as well as lindane in north eastern states of India [33-36]. The technical mixture of HCH were produced and used in India until it was banned in 1997, whereas lindane formulation are registered for use in public health practices to control vector borne diseases and for pest control in selected crops [37]. India is permitted to use DDT (10,000 t/year) under the Stockholm Convention, until an alternative can be found to combat vector borne diseases. During 2006-07, India used 6000 and 2560 MT of DDT for control of malaria and Kala Azar (*Leishmaniasis*), respectively [38].

Table 4: Compositional Isomer ratios of HCH, DDT, DDE and DDT compounds

Ratio	α/γ-HCH	DDT/DDE	DDE+DDD/∑DDT	DDTs/∑DDT	*o,p'/p,p'*-DDT	*p,p'*-DDT/∑DDT
Range	0-5.69	0-10.75	0-6.32	0-1.00	0-5.91	0-1.00
Mean	0.76	1.97	1.17	0.50	0.83	0.30

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The hypothesis of input of DDT was elucidated by evaluating the pattern of individual DDTs. After the DDT applications, much of the DDT gets slowly converted to DDE and DDD under aerobic as well as anaerobic conditions, respectively [39-40], hence ratios between the DDT and DDE and DDD is often used as an indication of age (recent or historic) of the source of DDT [41]. The relatively high concentration of DDT than DDD and DDE in this study indicated either that there was minimal degradation of DDT or there has been recent input of technical DDT. The ratio of DDE+DDD)/∑DDT >0.5 is indicative for a long-term biotransformation of DDT to DDD and DDE, while a ratio of less than 0.5 may reflect recent inputs [42]. In addition, a ratio of DDT/DDE >0.5 may indicate recent input of DDT, and, in contrast, a ratio of <0.3 may reflect past inputs of DDT. The residence time of *p,p'*-DDT could be estimated using the ratio of *p,p'*-DDT to ∑DDTs and DDTs/∑DDT. The DDT/DDE ratio for technical DDTs was reported to be 0.80 [43].

Table 2: Concentration of chlorinated pesticides in radish, spinach and tomato (µg·kg⁻¹ wet wt)

Compound name	Radish		Spinach		Tomato	
	mean±SE	range	mean±SE	range	mean±SE	range
α-HCH	0.93±0.23	<0.01-1.53	0.12±0.06	<0.01-0.24	0.67±0.34	<0.10-1.55
γ-HCH	1.57±1.03	<0.01-6.61	0.81±0.27	<0.01-1.15	1.20±0.63	<0.20-3.64
∑HCH	2.49±1.17	0.30-8.14	0.93±0.32	<0.01-1.39	1.87±0.59	<0.30-3.79
DDT	5.13±2.10	<0.01-12.09	0.55±0.39	<0.10-1.72	1.16±0.63	<0.10-3.40
DDD	2.52±0.96	<0.01-11.33	0.14±0.03	<0.10-0.23	0.34±0.18	<0.10-1.05
DDE	0.87±0.50	<0.01-3.28	0.56±0.22	<0.01-1.06	0.11±0.03	<0.01-0.22
∑DDT	8.52±3.96	<1.10-25.68	1.24±0.61	<0.20-3.00	1.61±0.80	<0.30-4.50
Aldrin	1.31±0.78	<0.01-5.04	0.30±0.14	<0.01-0.64	0.33±0.25	<0.01-1.31
Dieldrin	5.79±1.58	<0.01-34.11	0.07±0.02	<0.01-0.14	0.08±0.02	<0.01-0.13
Heptachlor	5.88±2.44	<0.01-13.16	0.73±0.36	<0.01-1.36	1.01±0.35	<0.20-2.22
∑OCPs	**24.00±6.60**	**<2.40-75.08**	**3.28±1.30**	**<0.20-6.14**	**4.90±1.62**	**<1.60-10.49**

DDT, DDE and DDD are sum of o,p' and p,p' isomers

Table 3: Concentration of chlorinated pesticides in eggplant, cauliflower and carrot (µg·kg⁻¹ wet wt)

Compound name	Eggplant		Cauliflower		Carrot	
	mean±SE	range	mean±SE	range	mean±SE	range
α-HCH	0.25±0.13	<0.10-0.62	0.62±0.15	<0.20-1.24	0.48±0.11	<0.30-0.64
γ-HCH	0.77±0.50	<0.10-2.24	1.63±0.53	<0.20-3.31	0.72±0.09	<0.60-0.84
∑HCH	1.02±0.50	<0.10-2.43	2.25±0.58	<0.40-3.83	0.60±0.36	<0.01-1.48
DDT	0.34±0.05	<0.20-0.43	1.05±0.30	<0.01-2.05	1.12±0.73	<0.10-3.27
DDD	0.17±0.04	<0.10-0.27	0.37±0.14	<0.01-1.01	0.48±0.25	<0.10-1.19
DDE	0.20±0.14	<0.01-0.63	3.09±0.94	<0.40-6.29	0.15±0.03	<0.10-0.19
∑DDT	0.78±0.15	<0.40-1.14	4.51±0.70	<2.20-6.49	1.67±0.96	<0.30-4.46
Aldrin	0.24±0.12	<0.01-0.59	0.79±0.19	<0.10-1.31	0.83±0.35	<0.30-1.33
Dieldrin	0.10±0.02	<0.01-0.16	0.13±0.02	<0.10-0.16	0.09±0.03	<0.01-0.15
Heptachlor	1.05±0.20	<0.01-1.39	3.00±1.02	<0.90-7.81	1.12±0.63	<0.20-2.01
∑OCPs	**2.92±0.70**	**<0.10-4.14**	**10.53±1.35**	**<7.30-16.48**	**3.30±1.01**	**<0.30-4.53**

DDT, DDE and DDD are sum of o,p' and p,p' isomers

The ratio of *p,p'*-DDT/∑DDTs, DDTs/∑DDT, DDE+DDD)/∑DDT, and DDT/DDE in this study was 0.30, 0.50, 1.17 and 1.97 respectively (Table 4), which indicates that these areas have not been sprayed with DDTs more recently and DDT contaminations were from biotransformation and transported depositions. The ratio of *o,p'*-DDT/*p,p'*-DDT can be used to distinguish technical DDT from "Dicofol-type DDT". The *o,p'*-DDT/*p,p'*-DDT ratio was reported to be 0.2~0.26 in technical DDT and 7.5 in dicofol products. In our study the ratio of *o,p'*-DDT/*p,p'*-DDT is 0.83, which is

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different from the scenario in China where dicofol usage is a major source of DDT [35]. Thus, the DDT contaminations in this region may be from technical DDT and not from dicofol usage. The ratios between parent DDT and metabolites in present study are indicative of biotransformation and recent input of DDT to the environment.

Aldrin/dieldrin was largely used as agricultural insecticide and for the control of tsetse fly, the vector of human and animal trypanosomiasis. Heptachlor (CAS No. 76-44-8) is a chlorinated dicyclopentadiene insecticide that tends to persist in the environment and accumulates in the food-chain. Although, Aldrin/dieldrin and heptachlor has been banned for manufacturer, use, import and export in India, these pesticides are still persistent in environment [44-46] and in biological samples where these may be associated with adverse effects [47-49].

Comparison of OCPs in Vegetables with other regions
A comparison of results of this study was made with other reports to assess the status of OCP contamination in vegetables from Kolkata market. The data of this study shows that the $\sum$HCH levels in vegetables in this study were lower than vegetables from Kanpur, India [9], Tianjin, China [15], Agra, India [50], Meerut, Muzaffarnagar and Ghaziabad districts of Uttar Pradesh, India [27], Haryana, India [51], and, cities of Central Uttar Pradesh, India [19]. However, HCH concentrations were higher than vegetables from Deyang and Yanting, [17] and Shanghai, China [52]. A much higher contamination of vegetables with $\sum$HCH compared with a MRL of 50 µg kg^{-1} was reported in vegetables from Kanpur, India, Hissar, India, and Tianjin, China.

$\sum$DDT concentrations in Kolkata vegetables were higher than those in vegetables from Haryana (India), Deyang, Yanting, and Shanghai (China), however the concentrations were lower than those in vegetables from Kanpur, Agra (India), Tianjin (China), Agra, India except radish, where DDT were less than radish from Kolkata, Meerut, Muzaffarnagar and Ghaziabad district of Uttar Pradesh, India, Central Uttar Pradesh, India.

Concentration of Aldrin, dieldrin and heptachlor in vegetables from this study were lower than reported from Meerut, Muzaffarnagar and Ghaziabad district of Uttar Pradesh, India [27], Central cities of Uttar Pradesh, India [19], and Jaipur, India [10].

The variations in levels of pesticide residues in vegetables grown on Indian soil are due to the disproportionate usage of pesticides in India and the amount of pesticide residue varies from one place to another. The states like Uttar Pradesh, Tamilnadu, Andhra Pradesh, Haryana and Karnataka have highest use of pesticides while on the other hand the states like Bihar, West Bengal, North eastern states have lowest use of pesticides. Out of total consumption of pesticides in India, only 13-14 percent share of pesticides were used in vegetables and fruits.

CONCLUSION

Presence of the OCPs in vegetables was observed which a matter of concern is. However, the vegetables from Kolkata, India generally never exceeded the MRLs indicating minimal risk to the consumers. The study indicates the use of lindane as well as technical formulation of the HCH in the study area and, contamination of these vegetables with DDTs. This may be due to transportation of pollutants from nearby human settlement areas, where pesticides used for public health aspects.

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Therefore, identification and elimination of contamination sources of OCPs in vegetables is recommended for the protection of human health.

Acknowledgement

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